

**NSF Panel on Light Source Facilities  
AMO Physics Applications:  
a fruitful partnership**

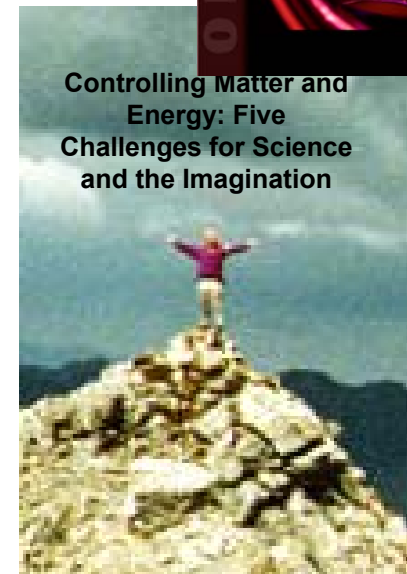
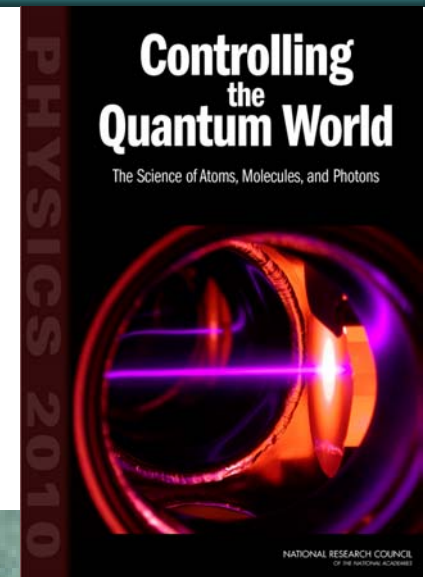
Philip Bucksbaum  
Stanford

Wednesday, January 10, 2008

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# Resources on AMO Research in 2010 and Beyond

- AMO 2010: Controlling the Quantum World
  - National Academy decadal survey
- Controlling Matter and Energy: Five Challenges for Science and the Imagination
  - Report from the Basic Energy Sciences Advisory Committee (BESAC)



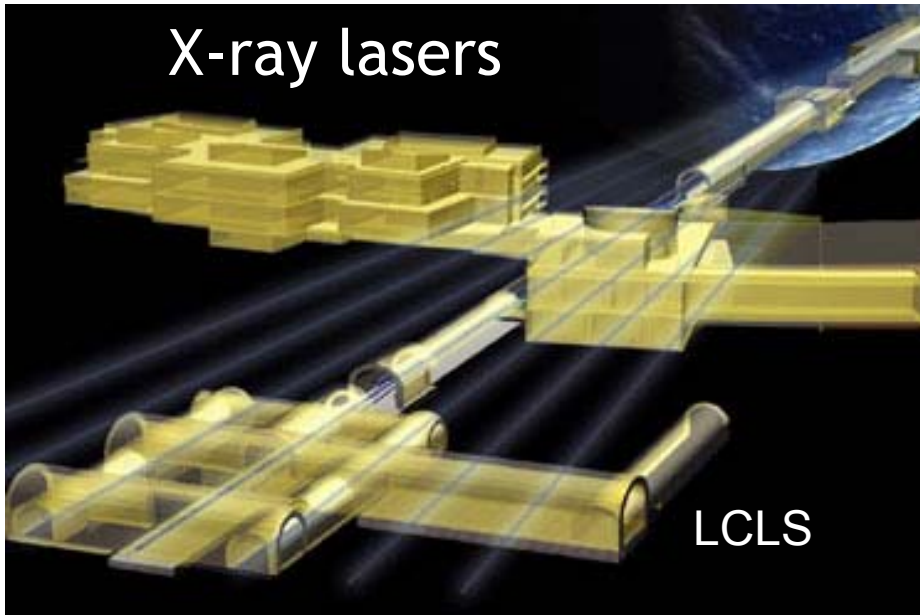
- High precision clocks – the precision time frontier.
- Imaging quantum processes on ultrafast time scales – the attoscience frontier
- Coherent molecular dynamics – the quantum control frontier
- Quantum computing and quantum communication: the quantum information frontier
- Atomic physics in exotic environments: the high field and high energy density frontiers
- Ultralow temperature phenomena: the frontier of quantum degenerate atomic gases

# Areas that make use of light source facilities

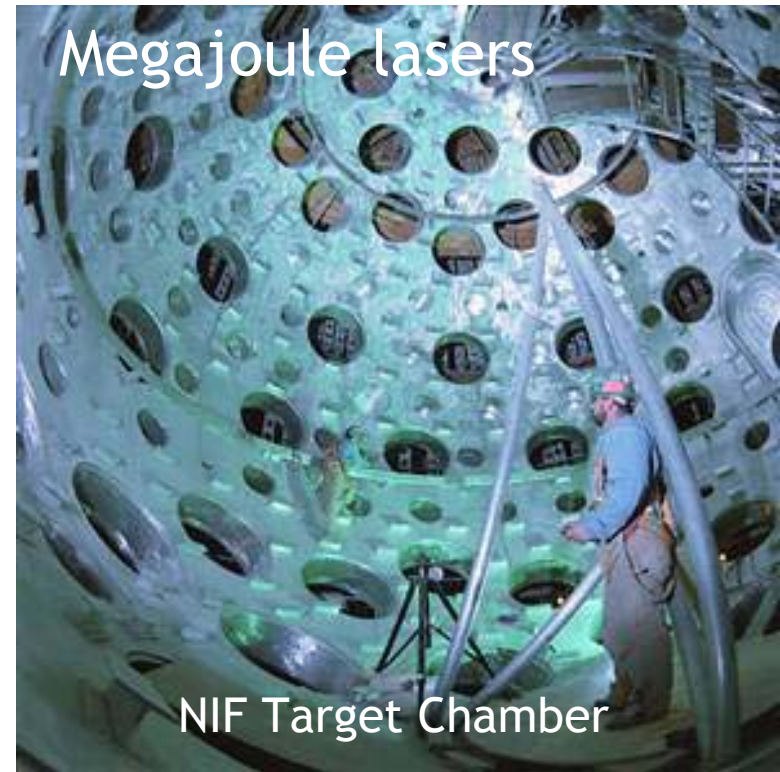
- High precision clocks – the precision time frontier.
- Imaging quantum processes on ultrafast time scales – the attoscience frontier
- Coherent molecular dynamics – the quantum control frontier
- 
- Atomic physics in exotic environments: the high field and high energy density frontiers
-

# The facilities of interest for AMO frontier physics

X-ray lasers



Megajoule lasers



Synchrotrons

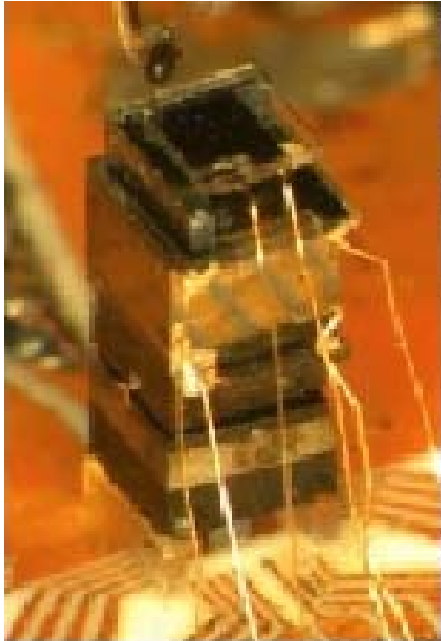
ALS



Smaller centers also  
contribute petawatt lasers  
for collaborative use  
(Texas Petawatt, FOCUS)

# Atomic Clocks

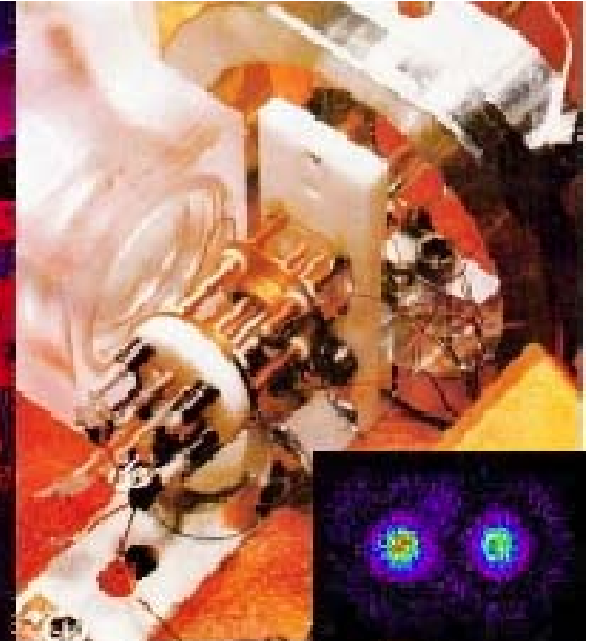
From military navigation to fundamental physics, atomic clocks are central.



Atomic clock in a  
1 mm<sup>3</sup> volume

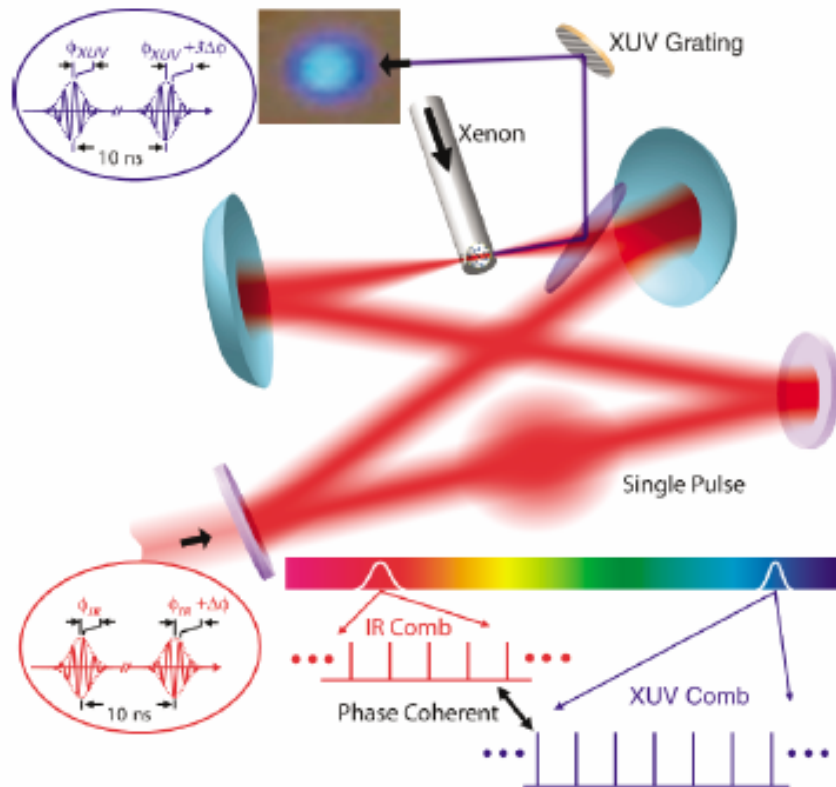


U.S. primary frequency standard: Cs  
atomic fountain clock, accurate to  
 $5 \times 10^{-16}$  or  
1 second in 60 million years



Optical frequency  
standards (Nobel '05) are  
the future of atomic clocks





Applications:

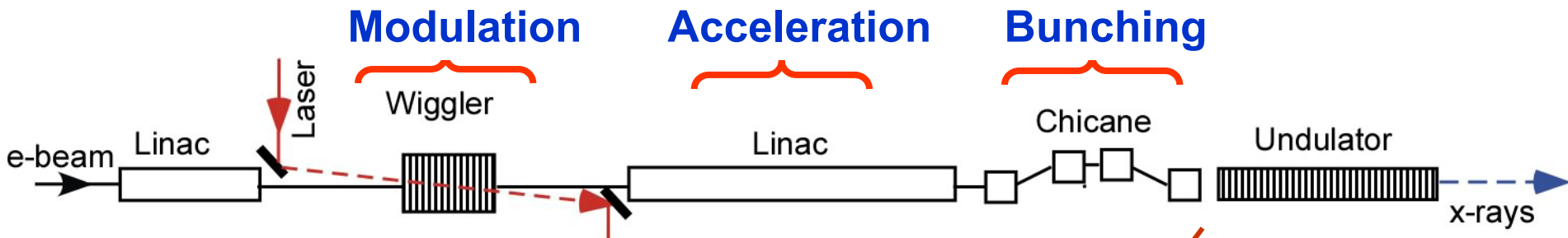
Precision vuv atomic spectroscopy

Optical time standard

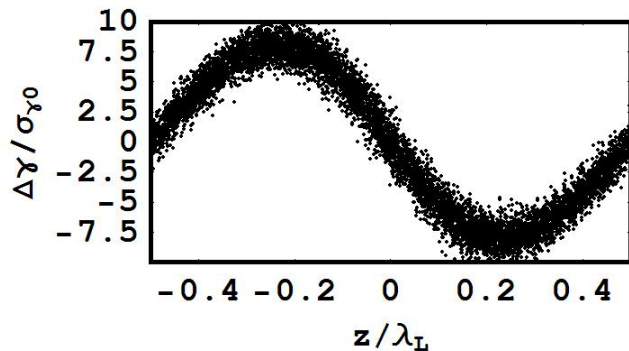
Time variations of fundamental processes

Jones et al. PRL 94, 193201 (2005)

# Connection II: Attosecond precision to tame the SASE FEL

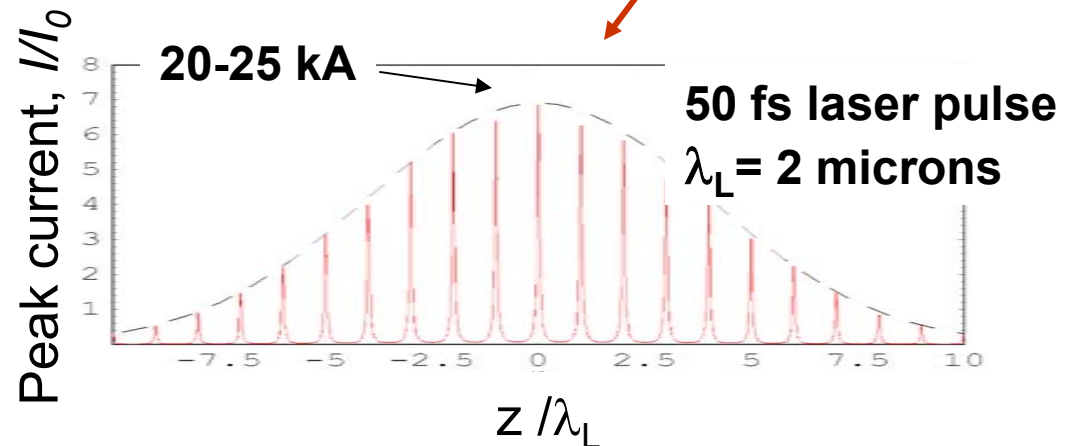


**Energy modulation in the wiggler at ~ 4 GeV**



Only one optical cycle is shown

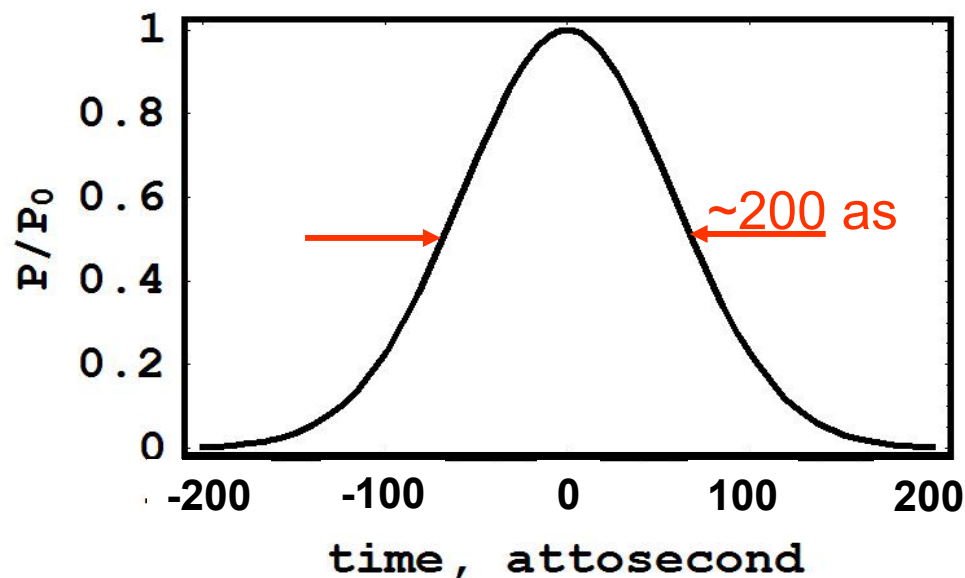
- Laser peak power ~ 10 GW
- Wiggler with ~ 10 periods



- Electron beam after bunching at optical wavelength

P. Emma, W. Fawley, Z. Huang, S. Reiche, G. Stupakov, A. Zholents

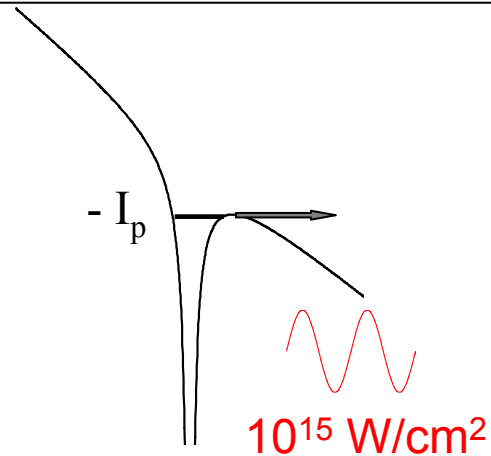




- Each spike is nearly temporally coherent and Fourier transform limited
- Carrier phase for an x-ray wave is random from spike to spike, but HHG injection schemes could overcome this
- Pulses less than 100 attoseconds may be possible with 800 nm laser

# High Field Frontier: Changing Strong Field Regimes Inside Atoms

**IR:**  
Low frequency regime



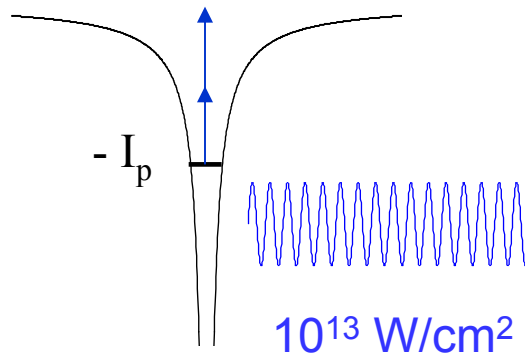
$$\Gamma_{e-A+} > \omega_{\text{laser}}$$

$$U_p > \hbar\omega_{\text{laser}}$$

$$\Gamma_{n\gamma \rightarrow e-A+} \approx \Gamma_{n+1\gamma \rightarrow e-A+}$$

ATI, HHG

**VUV FEL:**  
Intense photon source



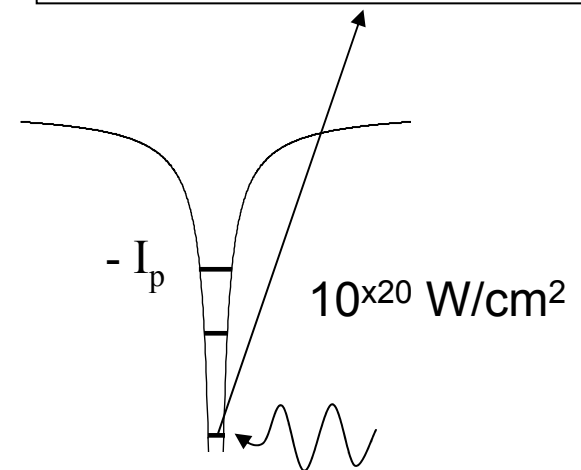
$$\Gamma_{e-A+} < \omega_{\text{laser}}$$

$$U_p \ll \hbar\omega_{\text{laser}}$$

$$\Gamma_{e-A+} \approx \Gamma_{2e-A++}?$$

sequential vs.  
non - sequential

**X-RAY FEL:**  
Highly ionizing source



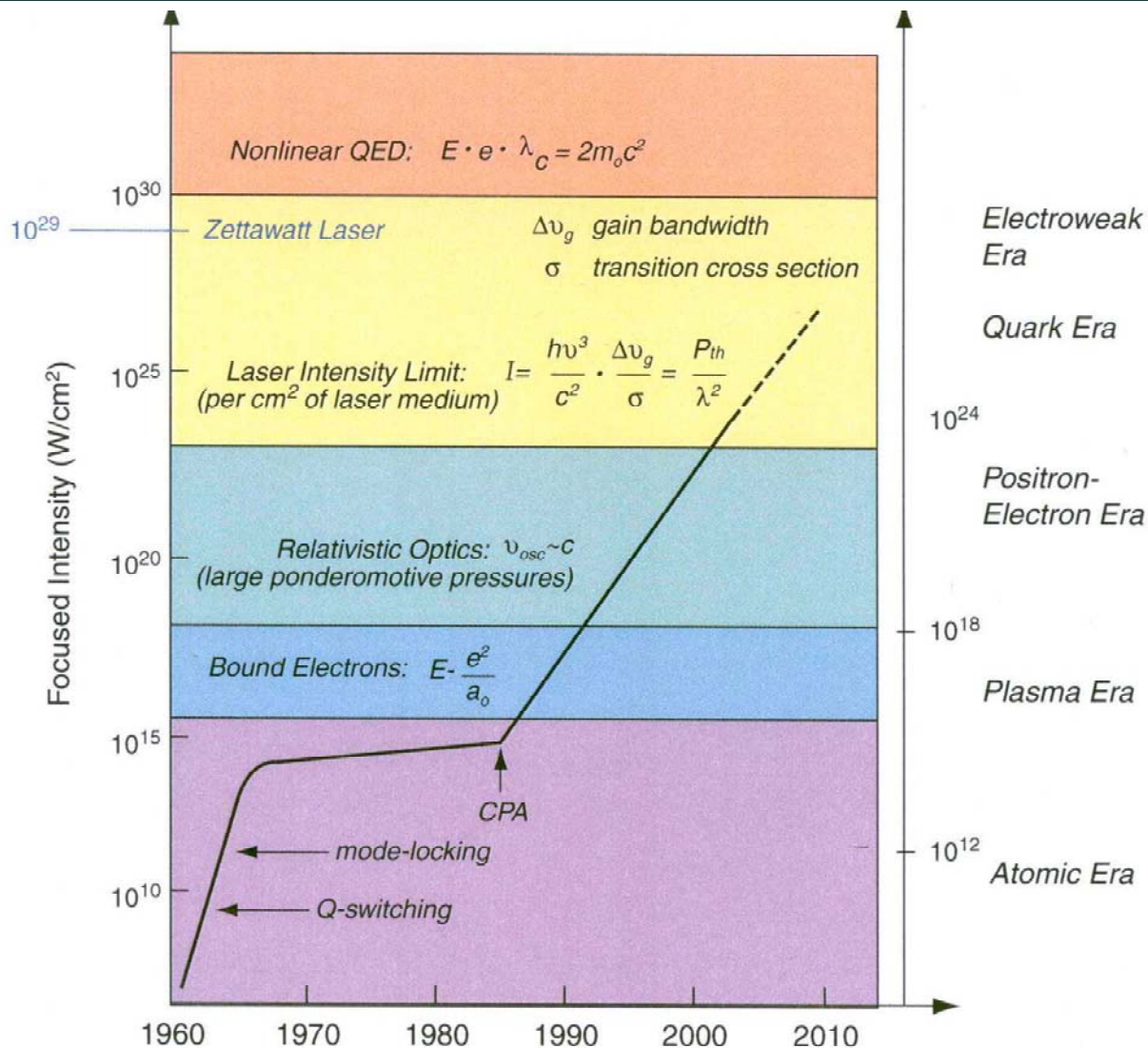
$$\Gamma_{e-A+} \ll \omega_{\text{laser}}$$

$$U_p \ll \hbar\omega_{\text{laser}}$$

$$\Gamma_{e-A+} \approx \Gamma_{\text{Auger}}, \Gamma_{2e-A++}$$

Hollow atoms?

# Laser Power Beyond a Petawatt can access Relativistic Strong Field Physics



Relativistic  
photoionization  
and rescattering  
of photoelectrons  
and ions

# Can the LCLS get to its own type of strong-field regime?

- impose a Keldysh parameter of one

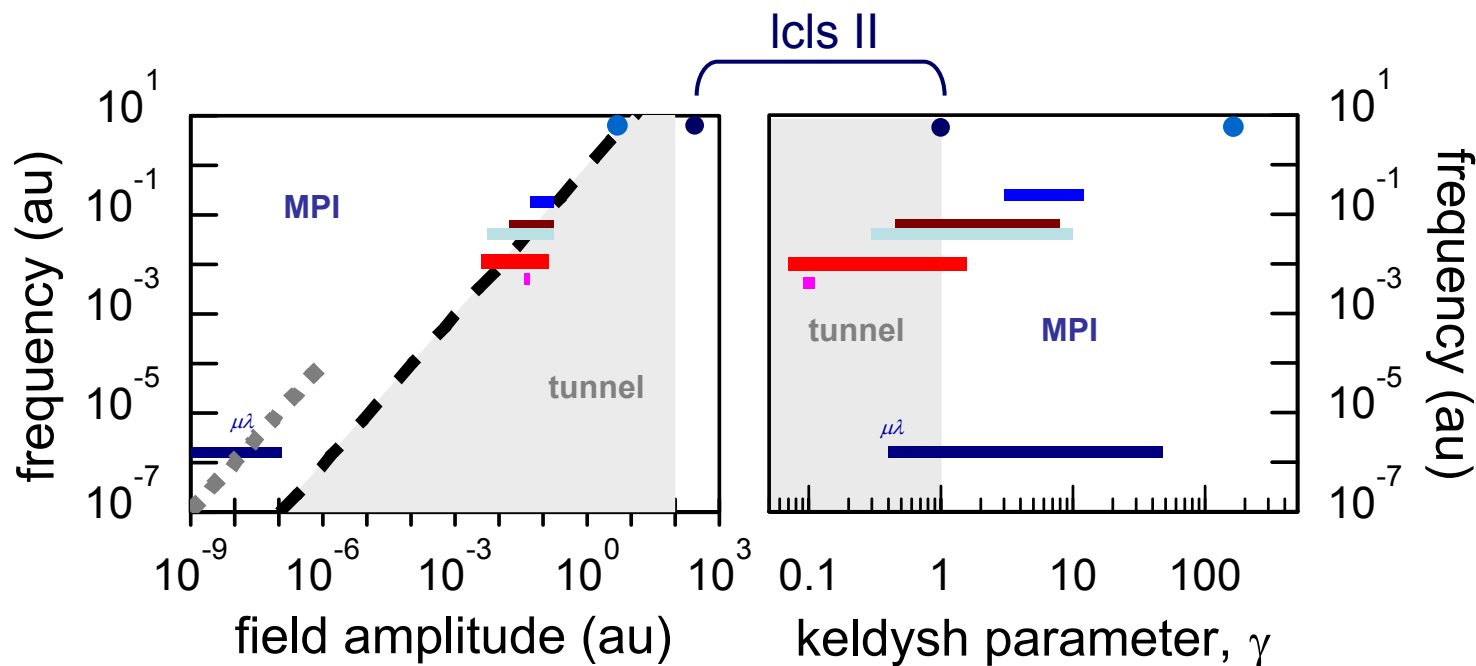
$$\gamma \equiv 1 = (I_p/2U_p)^{1/2} \Rightarrow U_p \cong 400 \text{ eV (8 eV)}$$

@ 800 eV, intensity needed is  $10^{21} \text{ W/cm}^2$  ( $10^{14} \text{ W/cm}^2$ )

number of photons is fixed, require tighter focus and shorter pulse

$\tau_{\text{lcls}} \sim 10 \text{ fs}$  (*very possible*)

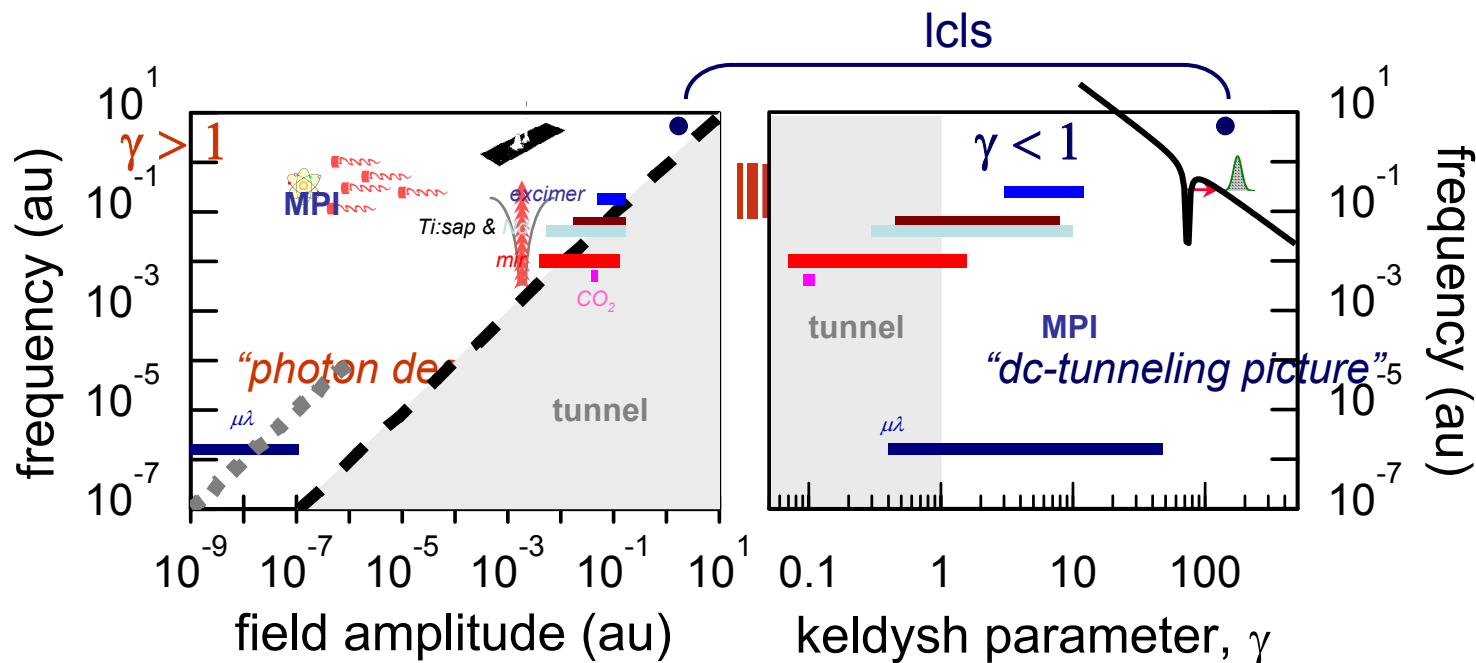
thus require a beam waist of 50 nm (in principle possible)



# Strong field wavelength scaling:

## Keldysh picture

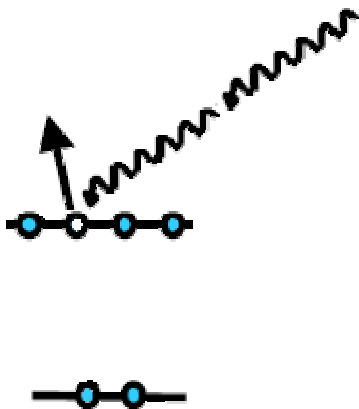
$$\gamma \equiv \frac{\text{optical frequency}}{\text{tunneling frequency}} = (I_p/2U_p)^{1/2} \propto \lambda^{-1}$$



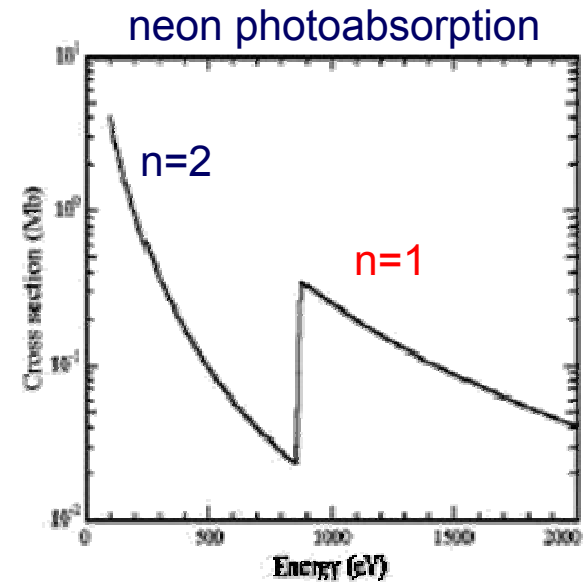
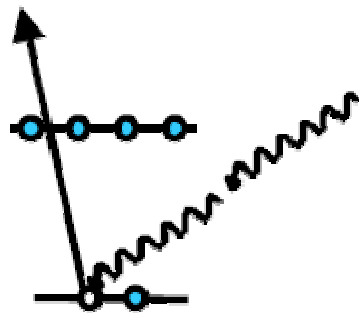
- data compiled based on both electron and ion experiments

# Physics comes from inner shells

- laser multiphoton ionization



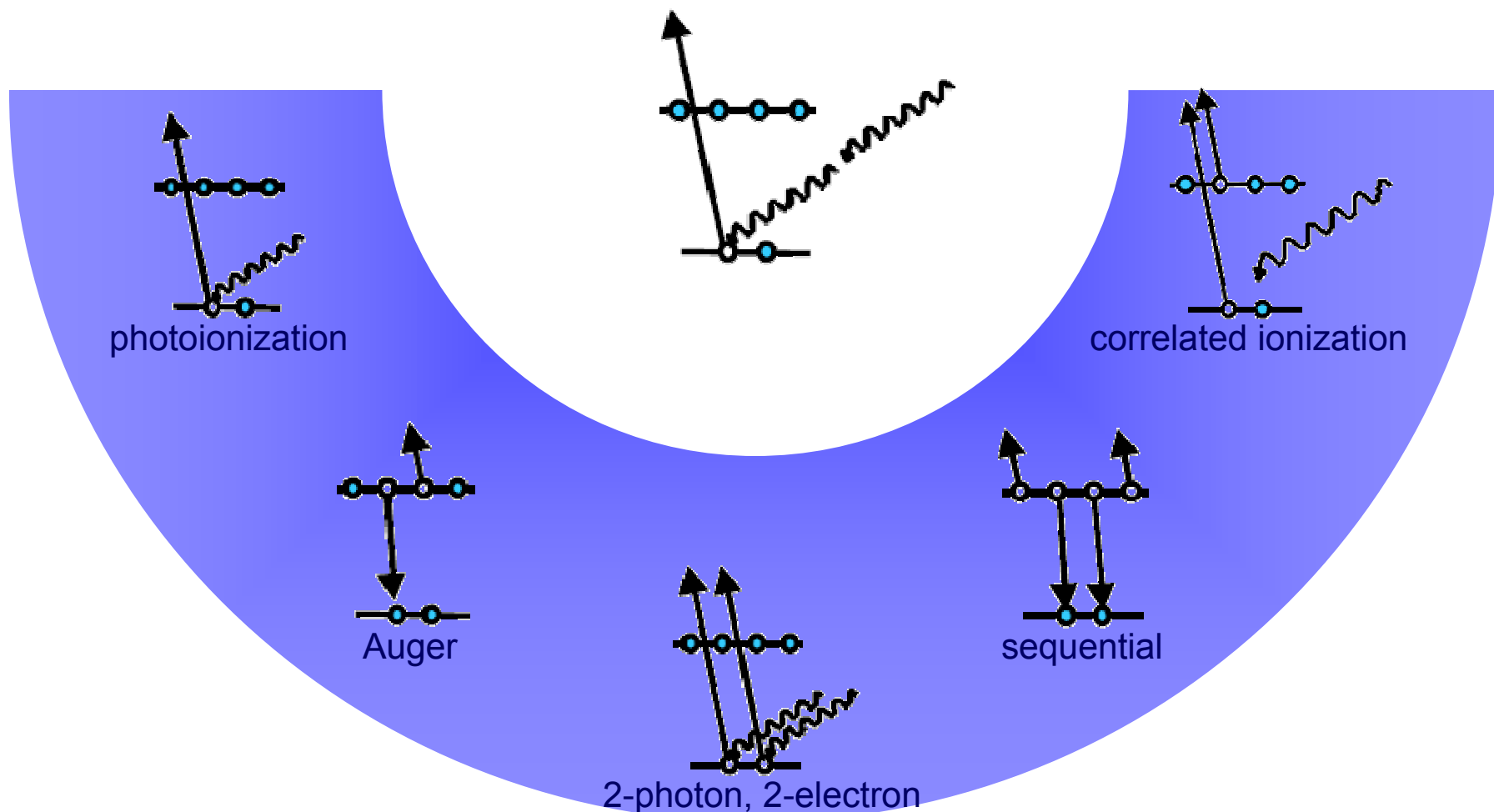
- x-ray multiphoton ionization





# x-ray strong field experiment

## x-ray multiphoton ionization



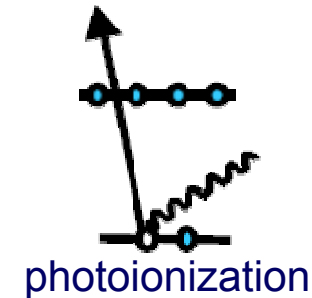
# 1-photon, 1-electron ionization

consider a 1-photon K-shell transition:

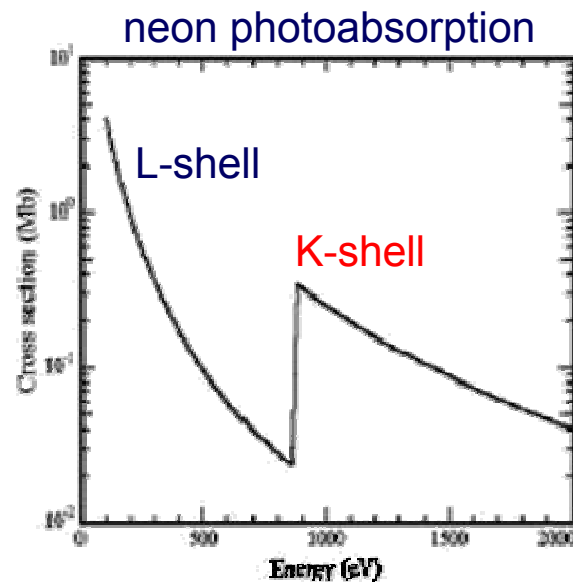
$$\sigma_K \approx 10^{-18} \text{ cm}^2$$

$$\Gamma_K = \sigma_K F_{\text{fclis}} \approx 10^{15} \text{ s}^{-1} \quad (\text{saturated})$$

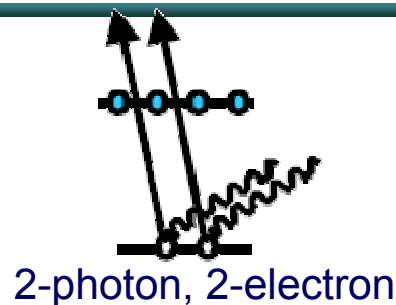
$$t_K = 1/\Gamma_K = 1 \text{ fs}$$



- rapid enough to ionize more than one electron!
- fast enough to compete with atomic relaxation?

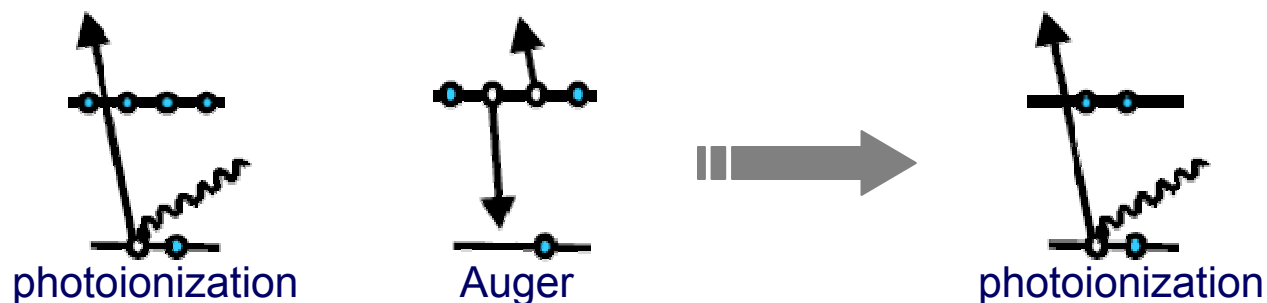


# 2-photon, 2-electron ionization

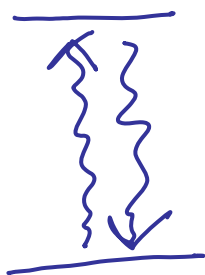


- are the electrons correlated?
- in a strong optical field single electron dynamics dominate.

i.e. is it sequential ionization?



- 3d generation x-ray sources have  $\ll 1$  photon per mode; LCLS will have coherence “spikes” containing a billion photons or more, in  $\sim$ femtoseconds! What about coherent excitation?



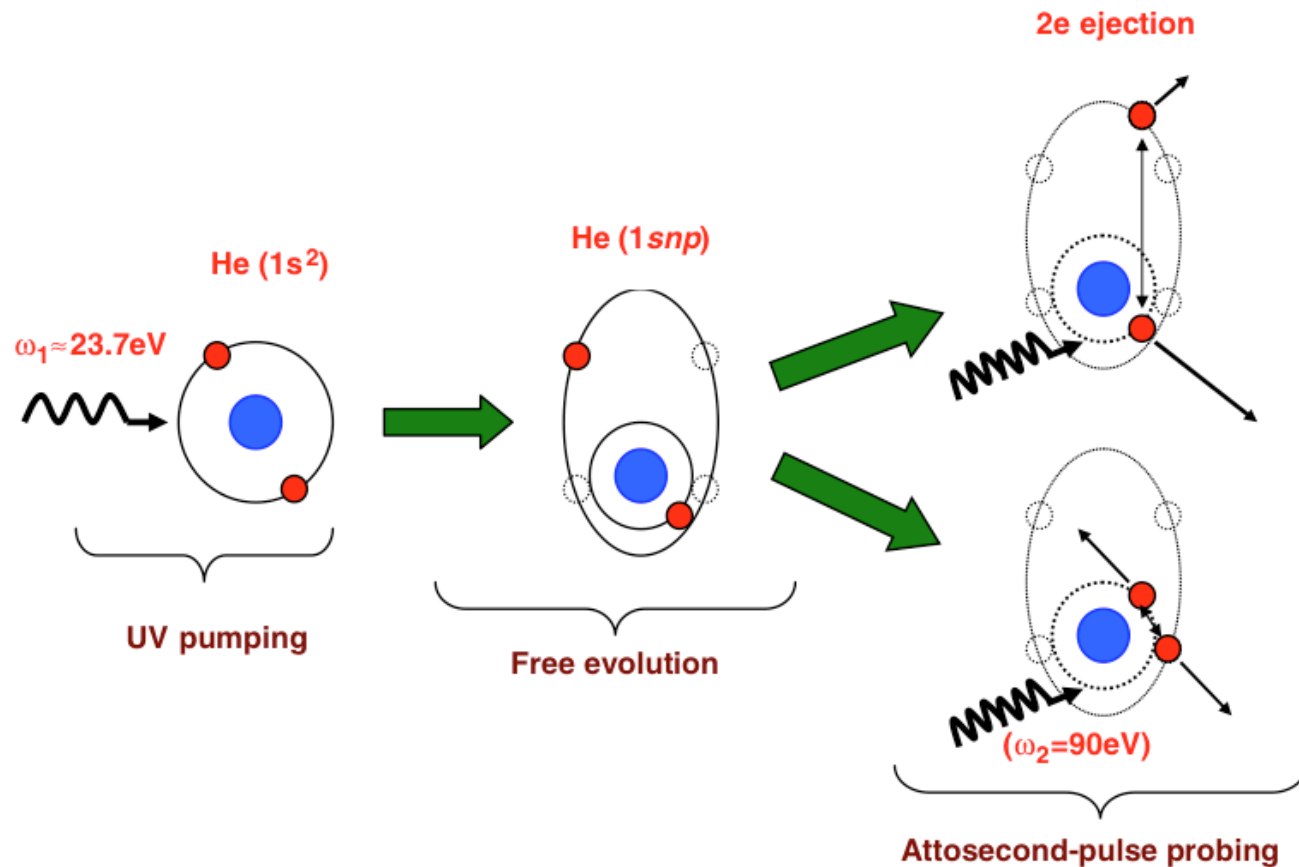
$$\text{Rabi rate } \Omega \approx \mu F_0$$

$$\mu = \langle f | ex | i \rangle \approx E_{\text{binding}}^{-1} \approx 0.01 \text{ for } 1\text{keV}$$

$$F_0 \approx 1 \text{ then implies } \Omega^{-1} \approx 2.5 \text{ fsec}$$

# Attosecond pump-probe?

fundamental time scale electron dynamics Atomic time unit = 24 attoseconds

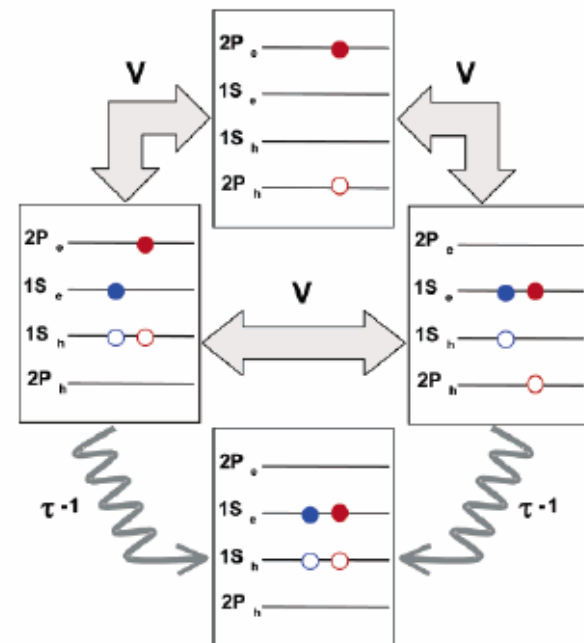
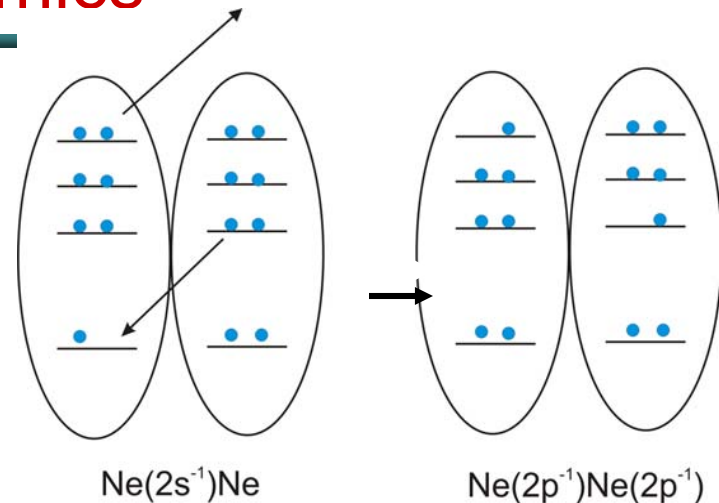


Direct attosecond probe of atomic electron correlation Hu and Collins, PRL (2006)

Theory

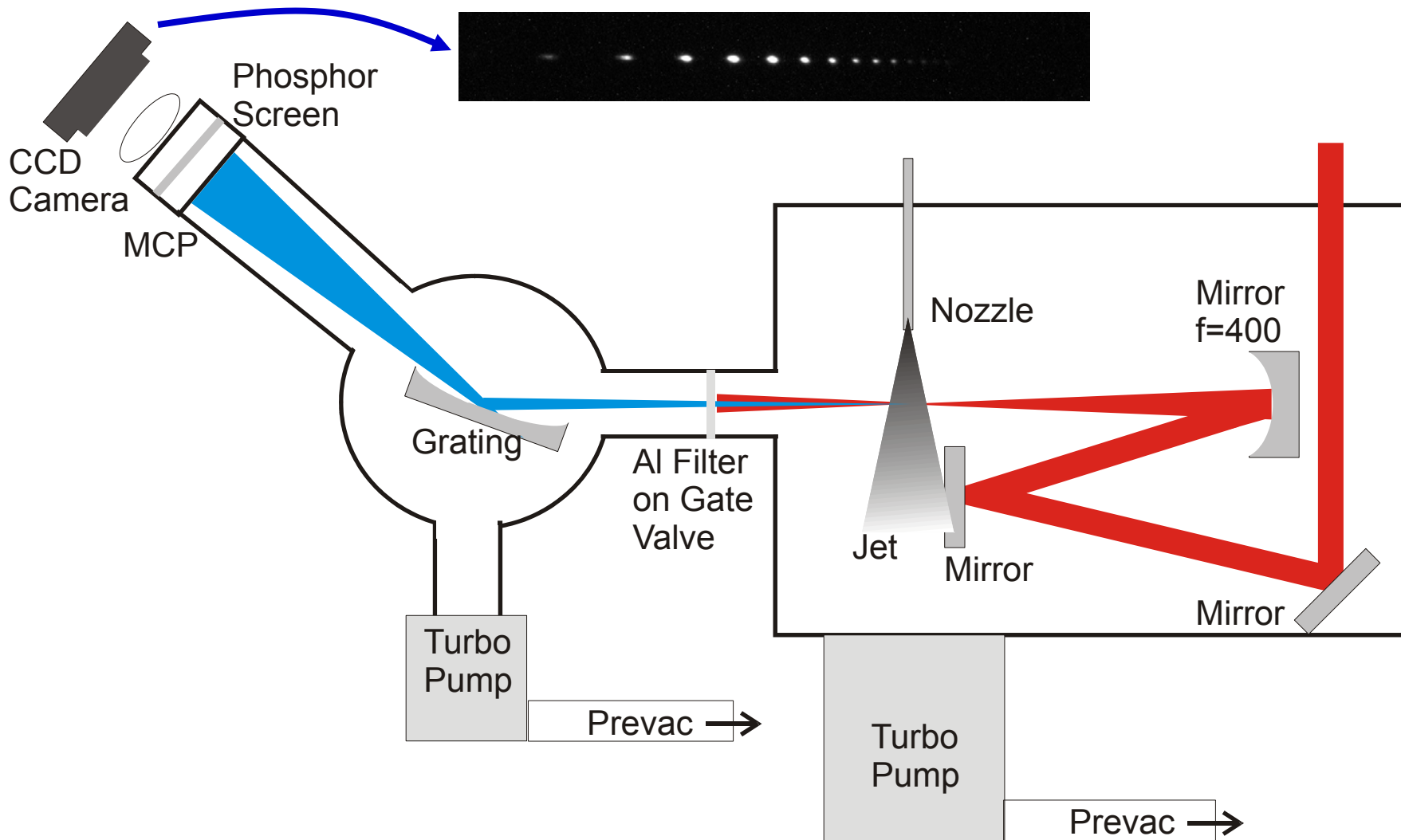
# Observing the inner workings of an atom: Auger dynamics

- Dynamics driven by electron correlation effects
- Time-scale important in atoms, molecules, nano-particles (i.e. multiple-exciton generation)
- Attosecond pump-probe expts proposed for NGLS (Berkeley workshop, 2007)

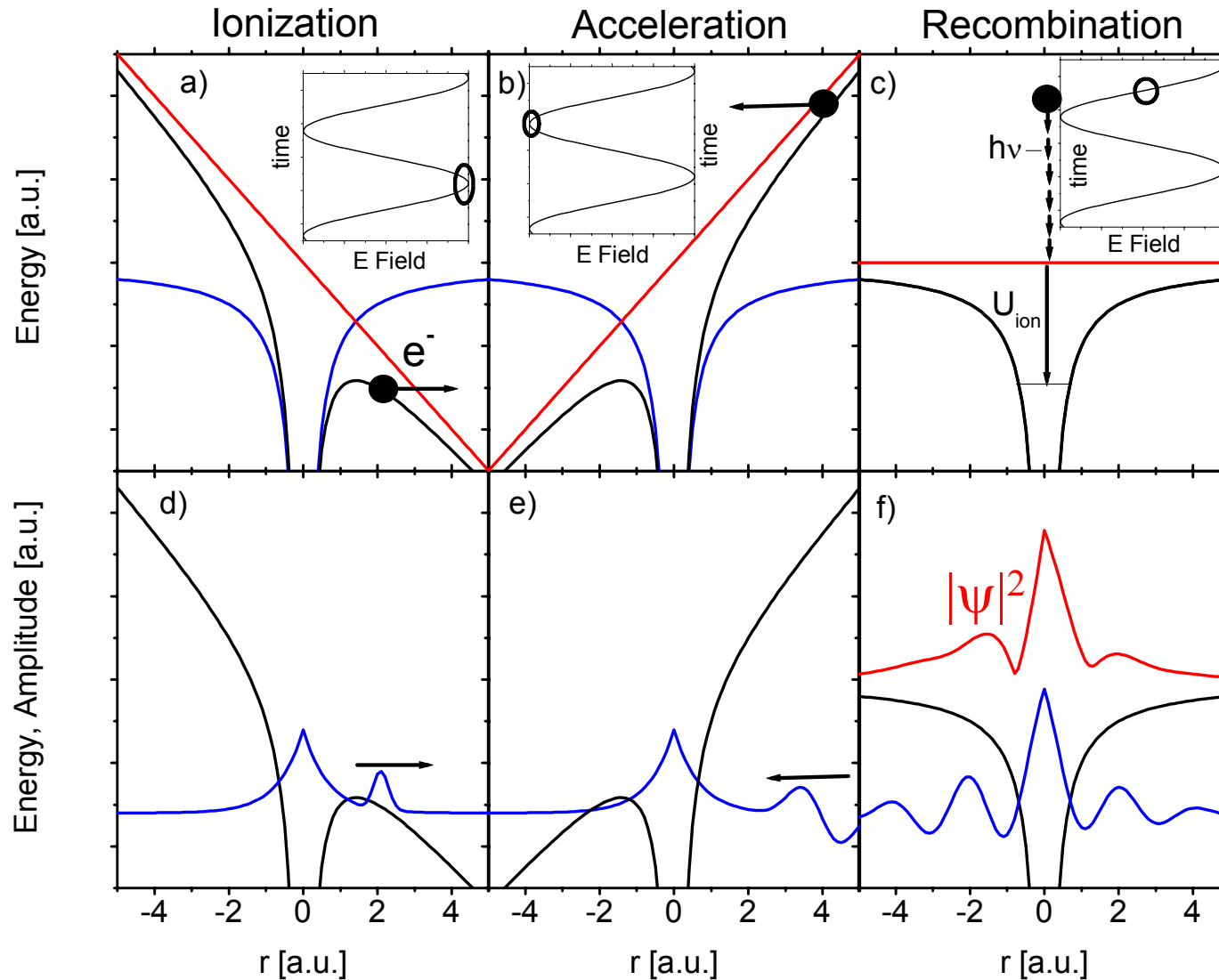




# The competition for attoscience: HHG, a source of controllable vuv radiation

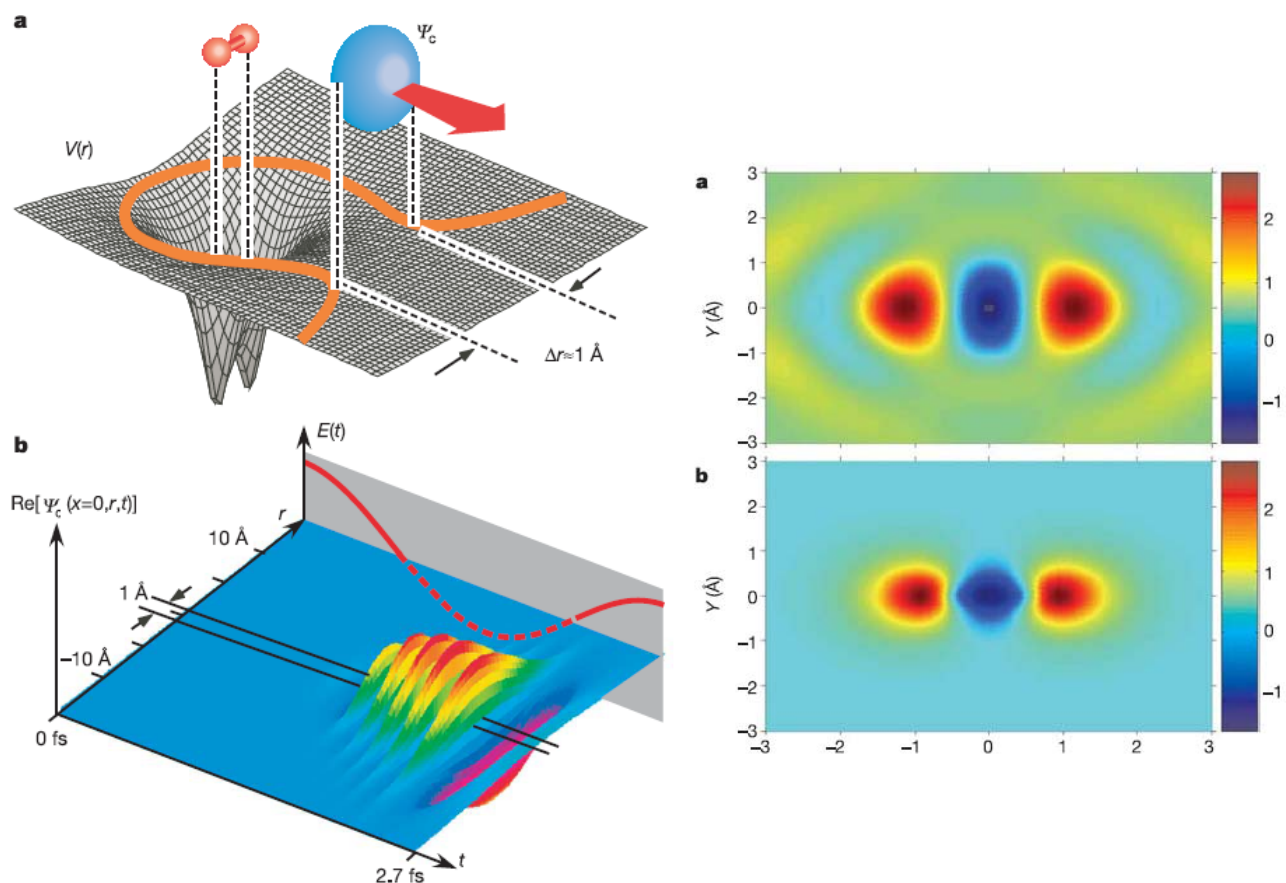


# HHG mechanism involves rescattering, and so is limited to the atomic scale



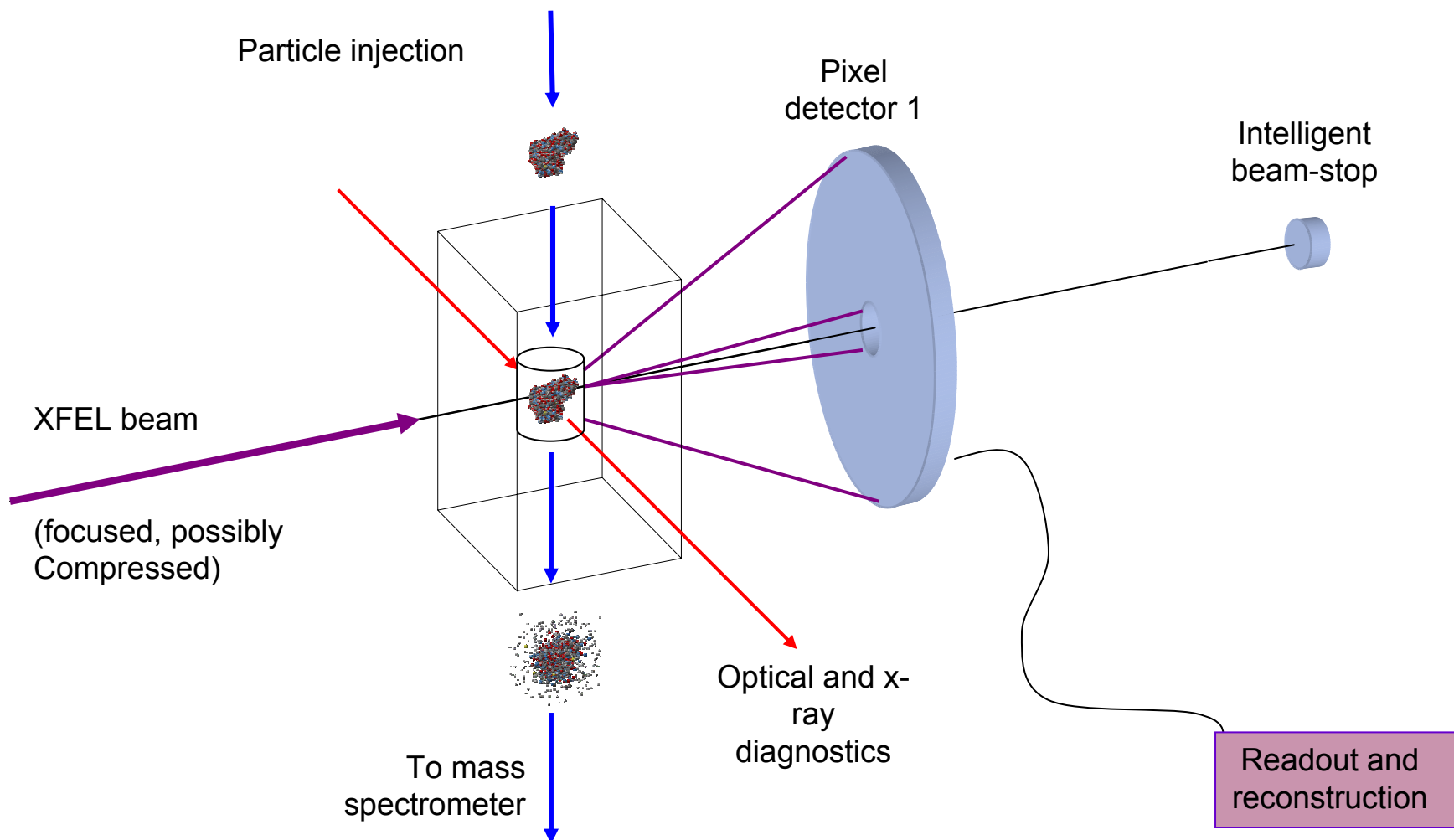
classical

quantum

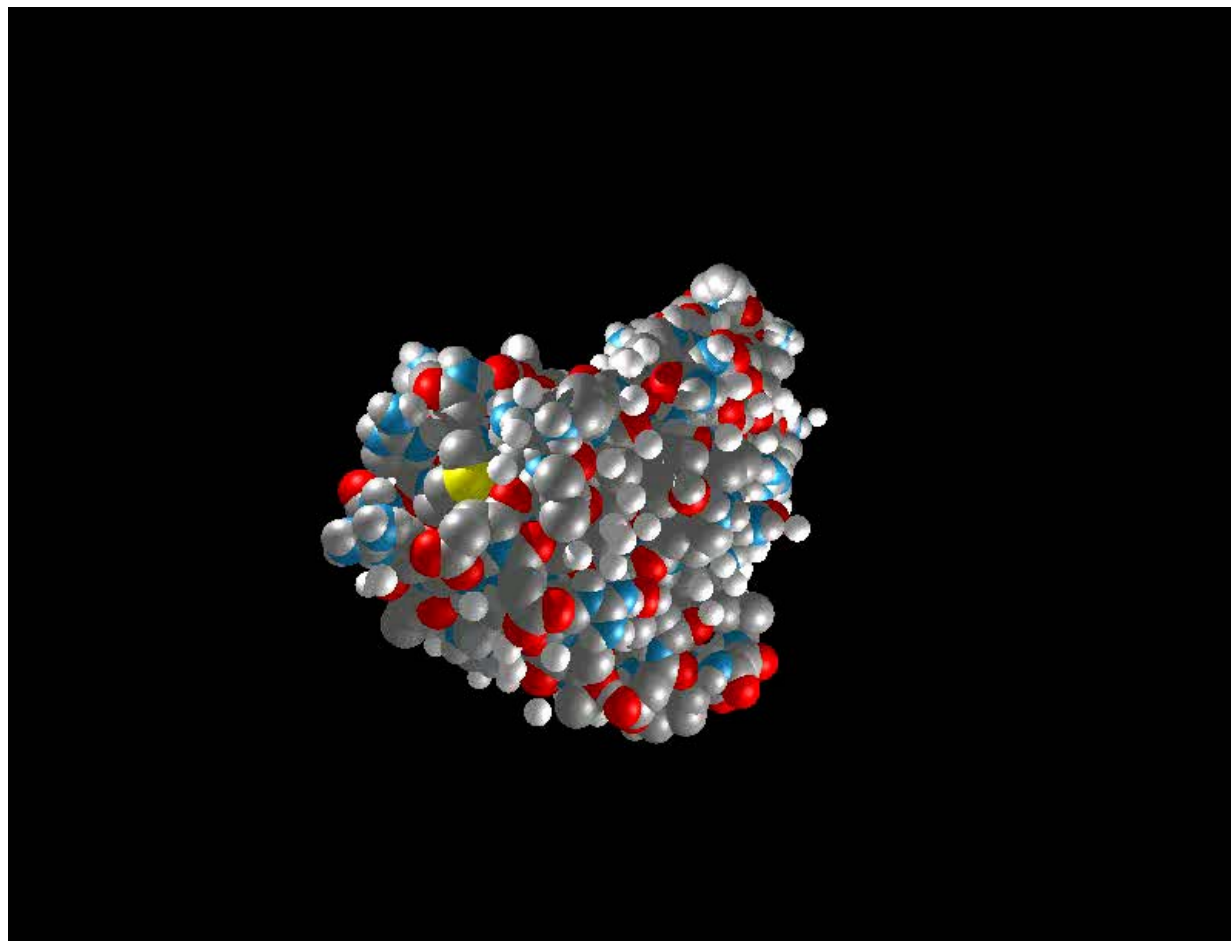


A snapshot image of a molecule  
 obtained from field ionization and electron-molecule recollision  
 Can we get this degree of control with radiation from a facility?

# Diffraction imaging with x-rays at LCLS



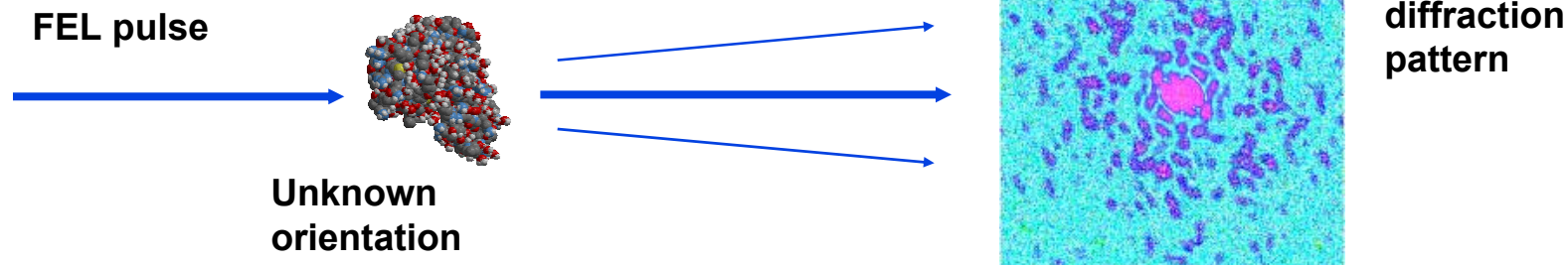
# Exploding T4 Lysozyme: Short pulses and high power are essential



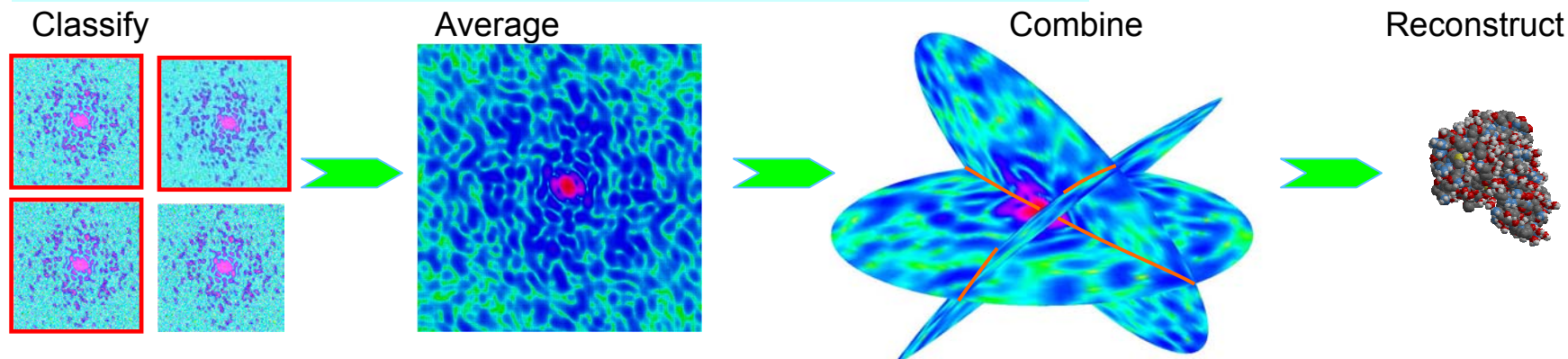
$1 \times 10^{11} \text{ W/cm}^2$  irradiation for 200 fs by LCLS pulse in 1mm spot (unfocused)

# A 3D dataset can be assembled from diffraction patterns in unknown orientations

## Diffraction from a single molecule:



## Combine $10^5$ to $10^7$ measurements into 3D dataset:



The highest achievable resolution is limited by the ability to group patterns of similar orientation

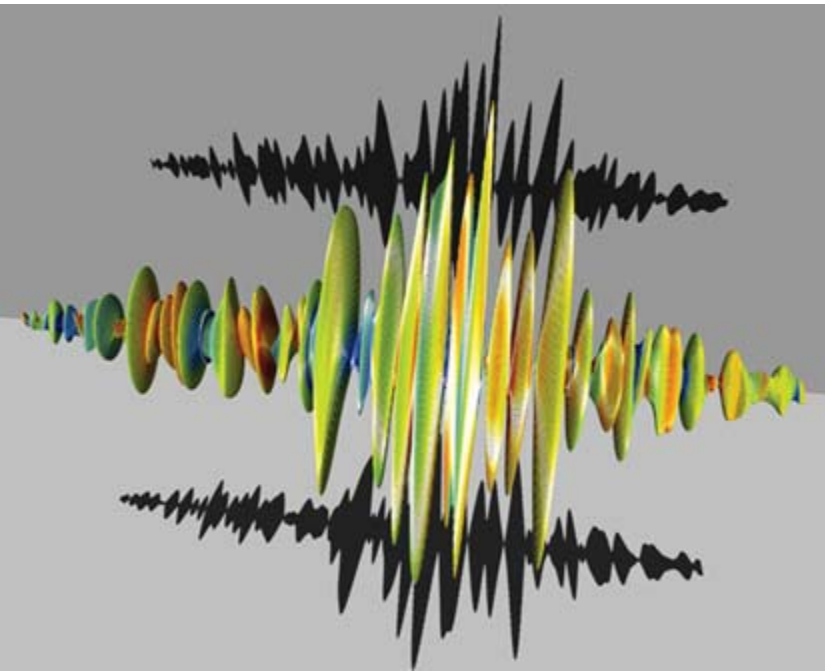
Gösta Hultdt, Abraham Szöke, Janos Hajdu (J.Struct Biol, 2003 02-ERD-047)

Miao, Hodgson, Sayre, PNAS 98 (2001)



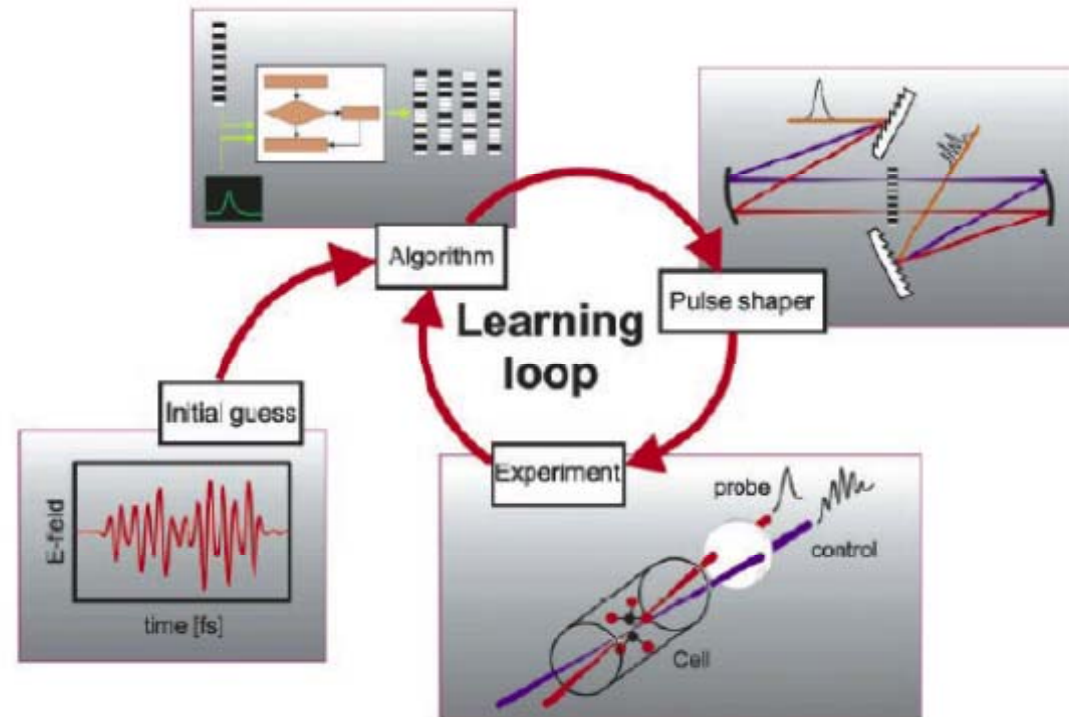
# Beyond imaging: Controlling quantum evolution

tools for control



Total field control

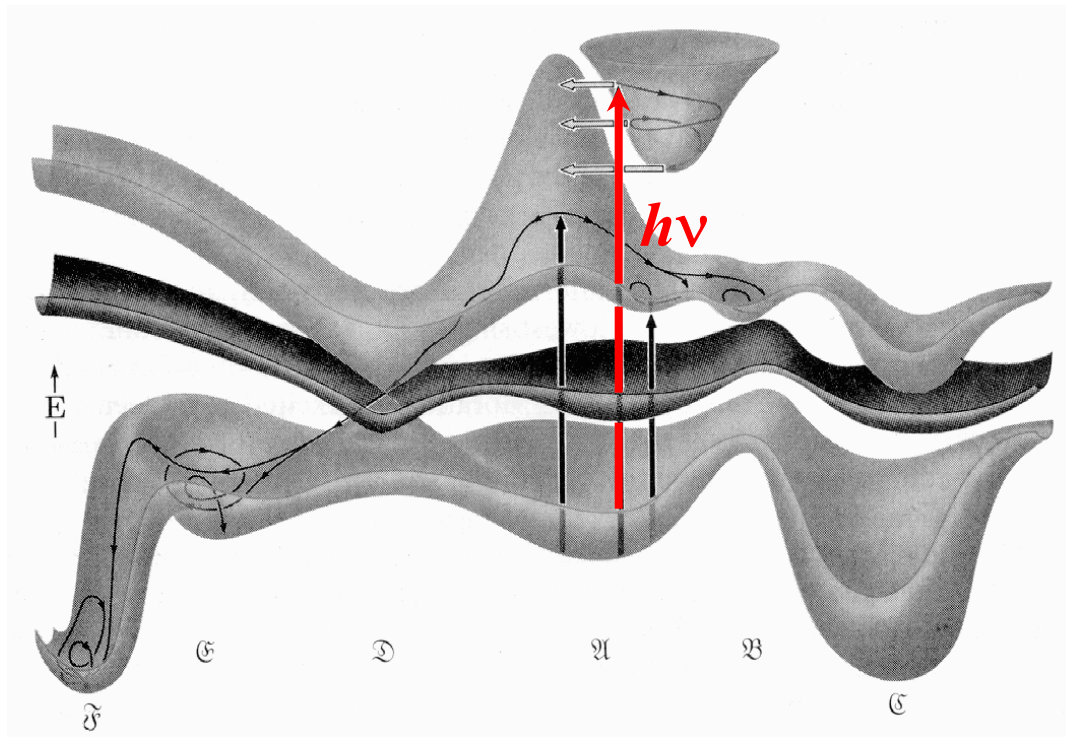
- Ultrafast studies (1999 Nobel)
- Carrier envelope phase control (2005 Nobel)
- Pulse shapers



Learning feedback

# Ultrafast vuv control will involve extreme non-Born-Oppenheimer dynamics

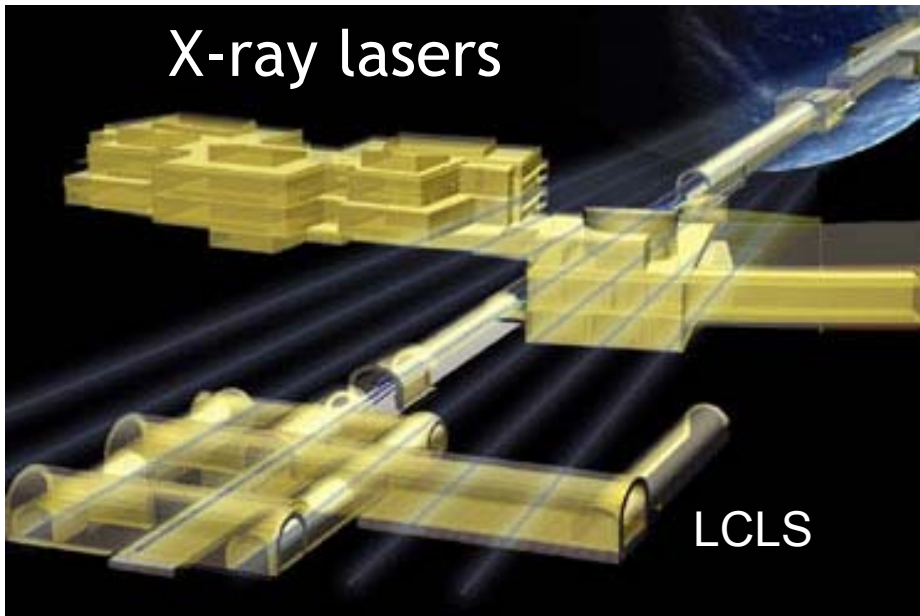
## Launch dynamics on multiple potential energy surfaces



Create electronic wavepacket: superposition on several electronic surfaces

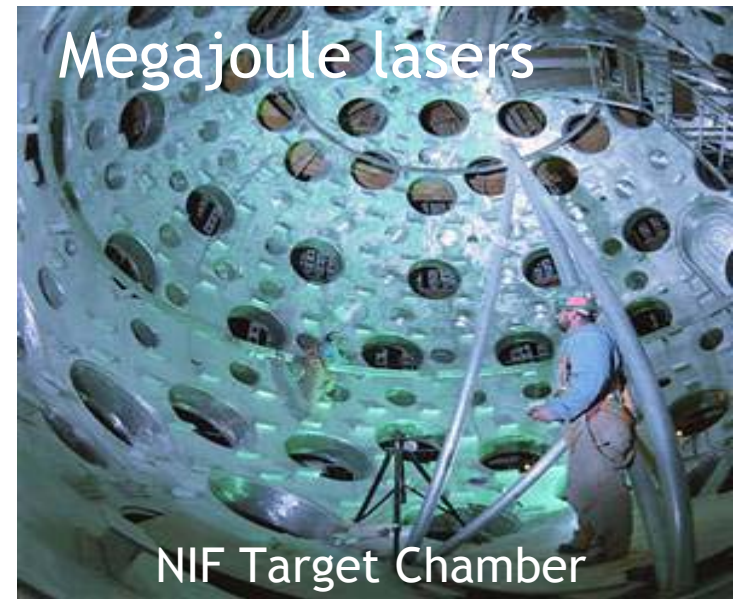
# Partnership: Light sources will contribute richly to AMO Science...

X-ray lasers



LCLS

Megajoule lasers



NIF Target Chamber

Synchrotrons

ALS



...and AMO will contribute techniques and technology to utilize these sources for science.

# Thanks to...

- Lou DiMauro
- Linda Young
- Markus Guehr
- Bill McCurdy
- Joe Stohr
- Janos Hajdu
- Many others...